Large thermomagnetic effects in weakly disordered Heisenberg chains

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The interplay of different scattering mechanisms can lead to novel effects in transport. We show theoretically that the interplay of weak impurity and Umklapp scattering in spin-1/2 chains leads to a pronounced dip in the magnetic field dependence of the thermal conductivity κ at a magnetic field $B \sim T$. In sufficiently clean samples, the reduction of the magnetic contribution to heat transport can easily become larger than 50% and the effect is predicted to exist even in samples with a large exchange coupling, $J \gg B$, where the field-induced magnetization is small. Qualitatively, our theory might explain dips at $B \sim T$ observed in recent heat transport measurements on copper pyrazine dinitrate, but a fully quantitative description is not possible within our model.

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Some of the most fascinating manifestations of quantum many-body physics in one-dimension (1D) can be found in spin-$\frac{1}{2}$ chain systems[1,2]. In particular, the spin-$\frac{1}{2}$ Heisenberg model with antiferromagnetic nearest neighbor exchange interactions is one of the most extensively studied paradigms. It provides a remarkably non–trivial example of an exactly solvable model[3], allowing a detailed analysis of its thermodynamic properties. While integrability is special to this particular model, its low energy properties are generic: they do not differ essentially from low energy properties of other (non integrable) spin chain models, with more general finite range interactions. In essence, these systems support Fermionic elementary excitations – spinons – which carry spin and no charge. Their kinetic energy and interactions are dictated by the exchange couplings in the chain, and their Fermi momentum can in principle be tuned by a magnetic field B, which plays the role of a chemical potential.

Thermodynamic properties of low-dimensional spin systems and especially of the Heisenberg model are generally very well understood allowing for a quantitative description of a broad range of experiments. In comparison, the heat and spin transport[4,5] in spin-systems is considerably more complicated. For example, the heat conductivity κ of the one-dimensional Heisenberg model is infinite at finite temperatures as the heat current operator is a conserved quantity for this idealized model. In real materials, the effects of disorder, phonon coupling and spin interactions not captured by the integrable Heisenberg model render the conductivity finite. Nevertheless, it remains often very large[6] as long as disorder is weak.

In this paper we study how the strong interactions of the Heisenberg model affect the heat transport in the system in the presence of weak disorder beyond the well known effect that disorder is strongly renormalized by the interactions[7]. Our study is directly motivated by recent experiments[5] in the spin-1/2 chain compound copper pyrazine dinitrate $Cu(C_4H_4N_2)(NO_3)_2$ (CuPzN) reproduced in Fig. 1. In this system a small exchange coupling, $J/k_B \approx 10.3\,K$, allows to polarize the system with moderate magnetic fields B. While both the spins and the phonons contribute to the heat transport at low temperatures T with similar strength, the field dependence can be used to separate the two effects. The total heat conductivity κ can be split into a contribution arising purely from phonons and a magnetic part, $\kappa(B,T) = \kappa_{\rm ph}(T) + \kappa_{\rm mag}(T,B)$. Then, one can use the fact that $\kappa_{\rm ph}(T)$ is practically independent of B to extract

$$\begin{aligned}\Delta\kappa_{\rm mag}(T,B) &= \kappa(T,B) - \kappa(T,B=0)\\ &= \kappa_{\rm mag}(T,B) - \kappa_{\rm mag}(T,B=0). \quad (1)\end{aligned}$$

In CuPzN $\Delta\kappa_{\rm mag}(T,B)$ shows a pronounced dip as a function of magnetic field for small B and T. Interestingly, the position of this dip scales linearly with the temperature, $g\mu_B B_{\rm min} \approx 3k_BT$, pointing to a simple underlying mechanism. A dip in $\kappa_{\rm mag}(B)$ has been observed in numerical simulations[8] of classical spin-chains coupled to phonons but this dip occurs at $B \sim J$ and does not scale with T.

Several processes can contribute to $\kappa_{\rm mag}$, the field dependent part of κ. First, there is a positive contribution from heat transported by the spin-chains. Second, the heat conduction of the phonons is reduced when phonons scatter off spin fluctuations. Third, there is a contribution from spin-phonon drag which is usually positive and can also become very large[9]. The positive sign of $\kappa_{\rm mag}$ in CuPzN suggests that the first and possibly the third mechanism are dominating.

Within a fermionic interpretation of the spin excitations, the magnetic field enters as a chemical potential while T determines the broadening of the Fermi surface. Therefore a likely interpretation of the experiment is that the characteristic dip arises when the Fermi surface moves away from the momentum $k_F = \pi/2a$, corresponding to a half-filled system with lattice spacing a. The commensurate filling for $B = 0$ is very special as it allows for Umklapp scattering: two spinon excitations

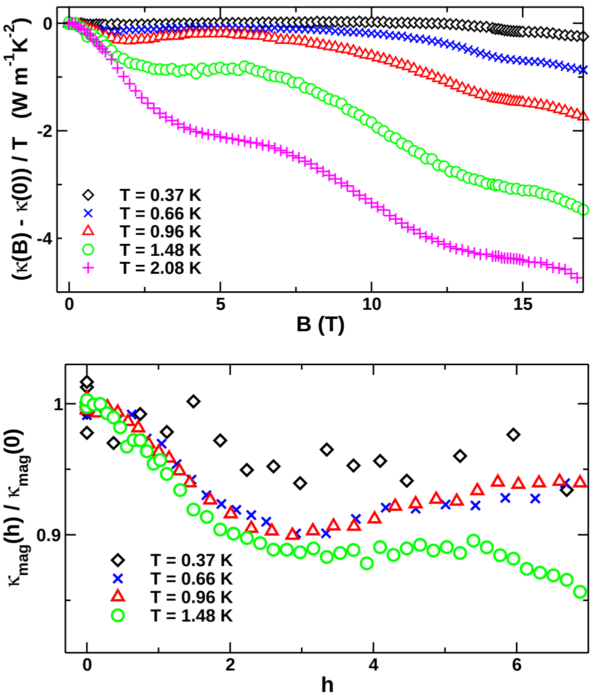

FIG. 1: (Color online). Upper figure: Field dependent thermal conductivity $\kappa(B,T)-\kappa(0,T)$ in the spin-1/2 compound CuPzN taken from Ref. [5]. The downturn at large fields arises as the spinon band is considerably depleted. For $T=0$ the magnetization saturates at $B\approx 15\,\mathrm{T}$. Lower figure: The normalized magnetic part of the thermal conductivity, $\kappa_{\rm mag}(B,T)/\kappa_{\rm mag}(0,T)$, as a function of $h=\mu_B gB/(k_BT)$ shows a dip at $h\sim 3$. Note that there might be a considerable error in the size of the dip as $\kappa_{\rm mag}(B=0,T)$ could not be measured directly due to a large phonon background, see text and Ref. [5] for details.

can be transferred from the left to the right Fermi surface and the excess momentum $4k_F=2\pi/a$ can be absorbed by the underlying lattice. As this scattering mechanism is only effective in the vicinity of the commensurate point, it is exponentially suppressed for $g\mu_B B>k_BT$. We will show that besides the Umklapp scattering it is necessary to include the effects of impurities to get structures at $B\sim T$; in the absence of disorder, the presence of certain approximate conservation laws[10] prohibits a relaxation of the heat current by the leading Umklapp process [see (5) below] alone.

Model and methods: We investigate the one-dimensional Heisenberg model in the presence of a magnetic field B and weakly disordered exchange couplings, $\delta J_i \ll J$

$$H \;=\; -\sum_i (J+\delta J_i)\,\mathbf{S}_i\cdot\mathbf{S}_{i+1} - g\mu_B B\sum_i S_z. \quad (2)$$

In the following, we will be interested in the limit of small magnetic fields and temperatures, $B,T\ll J$. More precisely, all calculations are done only to leading order in $1/\ln[(T,B)/J]$. The interactions lead to a strong renormalization of disorder[7], but we assume that the temperatures are sufficiently high, such that the renormalized disorder remains weak, $\delta J \ll \sqrt{JT}$.

For $B,T,\delta J_i\ll J$ one can use the powerful techniques of bosonization to describe the low-energy properties of the system. It is useful to split the effective low-energy Hamiltonian into three pieces

$$\begin{aligned} H &= H_{LL}+H_U+H_{dis} && (3)\\ H_{LL} &= v\int\frac{dx}{2\pi}\left(K(\partial_x\theta)^2+\frac{1}{K}(\partial_x\phi)^2\right) && (4)\\ H_U &= \frac{g}{(2\pi a)^2}\int dx[e^{i\Delta k x}e^{i4\phi}+h.c.] && (5)\\ H_D &= \frac{1}{2\pi a}\int dx\,\eta(x)[ie^{i2\phi}+h.c.] && (6)\end{aligned}$$

where (using the notation of [2,10]) $\partial_x\phi$ denotes fluctuations of the magnetization in the z direction, θ is the conjugate variable with $[\phi(x),\partial_x\theta(x')]=i\pi\delta(x-x')$, and we use units where $k_B=1,\hbar=1$. For the spin-rotationally invariant Heisenberg model, the velocity and the Luttinger parameters at the low-energy fixed point are given by $v=\frac{\pi}{2}Ja$ and $K=1/2$, respectively. The Umklapp term H_U describes the scattering of spinons from one Fermi point to the other. At a finite magnetization, the Fermi momentum $k_F=\frac{\pi}{2a}(1+2\langle S_z\rangle)$ deviates from $\pi/2a$ and therefore the excess momentum $\Delta k=4k_F-\frac{2\pi}{a}=4\pi\langle S_z\rangle/a$ cannot be absorbed by the crystalline lattice. For $\Delta k=0$ the Umklapp scattering is a marginally irrelevant operator whose strength decreases logarithmically with T (see below) while for $v\Delta k\gg k_BT$ it is exponentially suppressed in a clean system. The effects of disorder, or more precisely from components of the disorder potential oscillating with momentum $2k_F$, is described by H_D which models the scattering from one Fermi point to the other with $\eta(x)\sim\delta J(x)$. Here we assume uncorrelated disorder with $\langle\eta(x)\eta(x')\rangle=D_{dis}\delta(x-x')$.

To calculate the heat conductivity we use the so-called memory matrix formalism[11] as in [10]. Within this approach one calculates a matrix of relaxation rates for a given set of modes. As has been discussed in detail in [12], in general this method allows to calculate a lower bound to the conductivity. The formalism gives precise results as long as the relevant slow modes are included in the calculation. For the present system, the essential step is to realize[10,13] that in the absence of disorder the operator $Q=J_H+v\frac{\Delta k}{4K}J_s$ is conserved, $[Q,H_{LL}+H_U]=0$. Here, $J_H=v^2\int dx\,\partial_x\theta\partial_x\phi$ is the heat current associated with H_{LL} and $J_s=vK\int\partial\theta/\pi$ is the spin-current. This can be seen by realizing that up to the prefactor v^2 the heat current can be identified with the momentum operator, the generator of translations. The Umklapp term describes a process where a momentum Δk is generated and therefore its commutator with J_H is proportional to $v^2\Delta k$. Similarly, the spin current is changed by $-4v$ as two spinons with velocity v are scattered into states with velocity $-v$. Therefore the linear combination $Q=J_H+v\frac{\Delta k}{4K}J_s$ remains unaffected by Umklapp

processes. In terms of the original variables, Q can be identified with $J^2 \sum_i \mathbf{S}_i(\mathbf{S}_{i+1} \times \mathbf{S}_{i+2})$, the heat current operator for $B = 0$ which commutes with the integrable Heisenberg model (2) in the absence of disorder, $\delta J_i = 0$ (when longer range interactions or interchain coupling break integrability, the lifetime of Q nevertheless remains exponentially large in a clean system, see [10,13]). We therefore set up the memory matrix formalism in operator space spanned up by the two relevant currents J_H and J_s.

The decay rates of the currents are determined[10,11] from a 2×2 matrix $\hat{M}$ of correlation functions (the 'memory matrix'),

$$M_{ij} = \lim_{\omega\to 0} \frac{\mathrm{Im}\,\langle \partial_t J_i; \partial_t J_j\rangle_\omega}{\omega} \tag{7}$$

where $\langle A; B\rangle_\omega$ denotes a retarded correlation function of A and B evaluated at the real frequency ω and $J_1 = J_H$, $J_2 = J_s$ are the two relevant operators. Within the assumptions of our paper, we can treat both Umklapp and disorder perturbatively and therefore it is sufficient to evaluate all expectation values with respect to H_{LL} with $K = 1/2$ as the derivative $\partial_t J_i$ are already linear in the perturbations H_U and H_{dis}.

The heat conductivity per spin chain is obtained from

$$\kappa_{\mathrm{mag}} \approx \frac{\chi_H^2}{T}\, \hat{M}^{-1}\Big|_{11} = \frac{\chi_H^2}{T} \frac{M_{ss}}{M_{ss}M_{HH} - M_{sH}^2} \tag{8}$$

where $\chi_H = \langle J_H; J_H\rangle_{\omega=0} \approx \frac{\pi v T^2}{3}$ is the susceptibility of the heat current.

Separating the contributions from Umklapp and disorder, $\hat{M} = \hat{M}_U + \hat{M}_{\mathrm{dis}}$, we find using straightforward perturbation theory

$$\hat{M}_U = \Gamma_U(B,T) \begin{pmatrix} \left(\frac{v\Delta k}{2}\right)^2 & -\frac{v\Delta k}{2} \\ -\frac{v\Delta k}{2} & 1 \end{pmatrix} \tag{9}$$

$$\Gamma_U = -\frac{g(T)^2 v (\Delta k)^2}{8\pi^2} n_B'(v\Delta k/2) \tag{10}$$

$$\hat{M}_{\mathrm{dis}} = \frac{v\pi D_{\mathrm{dis}}}{8a} \begin{pmatrix} T & 0 \\ 0 & \frac{2}{\pi^2 T} \end{pmatrix} \tag{11}$$

where $n_B'(\omega)$ is the derivative of the Bose function, $n_B(\omega) = 1/(e^{\omega/T} - 1)$. The T dependence of $g(T) \sim \pi v/\ln(J/T)$ (see below) takes into account that the Umklapp scattering is marginally irrelevant with respect to the clean fixed point.

Results: The rather simple equations (8-11) describe the complex interplay of disorder and Umklapp scattering. First, in the absence of disorder, the heat conductivity is infinite[14] as $\hat{M}_U$ has an eigenvalue 0 reflecting the conservation of Q described above. Second, for vanishing magnetic field, $\Delta k = 0$, Umklapp scattering plays no role and one obtains the well-known result $\kappa_{\mathrm{mag}}/T \sim T$. The mean free path decreases linearly in T as the disorder is strongly renormalized by the interactions[7]. Third,

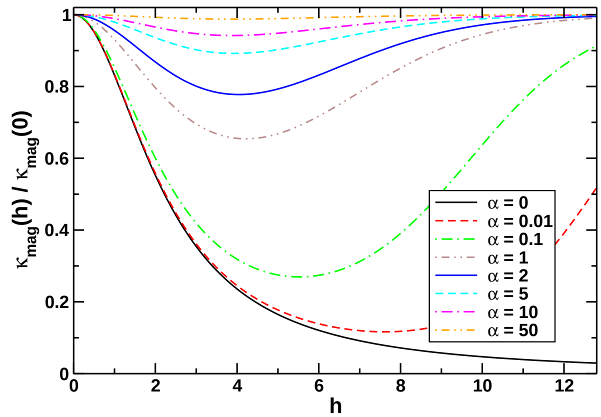

FIG. 2: (Color online). Field dependence of the of the normalized thermal conductivity as a function of the rescaled field, $h = g\mu_B B/k_B T$. For moderate disorder $\alpha(T)$, Eq. (12), $10 \gtrsim \alpha(T) \gtrsim 1$, a pronounced dip in $\kappa_{\mathrm{mag}}(B)$ is predicted for $g\mu_B B \sim 4k_B T$. For even smaller values of $\alpha(T)$ the dip gets broader and deeper, approximating the asymptotic behavior (14) (solid black line), up to larger and larger fields.

for finite magnetic field and $T \to 0$, Umklapp scattering is exponentially suppressed, $\Gamma_U \sim e^{-v\Delta k/2T}$, as the Fermi energy has shifted away from commensurate filling and one finds $\kappa_{\mathrm{mag}}(B \gg T) = \kappa_{\mathrm{mag}}(B = 0)$. Here we have neglected the – formally subleading – effect that the Luttinger liquid parameter K and therefore also the renormalization of the disorder potential depend on B.

Upon increasing the magnetic field B, the Fermi surface is shifted and Δk increases approximately linearly in B, $\Delta k \approx 4\pi\chi B/a$ with[15] $\chi \approx g\mu_B/(\pi^2 J)$. As argued above, for $\Delta k = 0$ the Umklapp scattering does not influence transport. Therefore, by raising Δk the effect of Umklapp scattering is switched on proportionally to $(\Delta k)^2$. But upon increasing Δk further, Umklapp scattering is switched off for $v\Delta k \gg T$ or $B \gg T$. The net result is a pronounced dip in $\kappa_{\mathrm{mag}}(B)$, see Fig. 2.

In the scaling limit of weak disorder and $1/\ln[(B,T)/J] \ll 1$ the normalized conductivity $\kappa_{\mathrm{mag}}(B,T)/\kappa_{\mathrm{mag}}(B = 0,T)$ is only a function of the scaling variable $h = \mu_B g B/k_B T$ and a dimensionless variable

$$\alpha(T) = \frac{D_{\mathrm{dis}} v^2}{(k_B T)^2 g(T)^2 a} \approx \frac{8 k_B v}{9\pi \tilde{g}(T)^2 \kappa_{\mathrm{mag}}(B = 0,T)} \tag{12}$$

which parameterizes the relative strength of (renormalized) disorder and Umklapp scattering ($\tilde{g} \sim 1/\ln[J/T]$ is defined below). In these variables, we obtain

$$\frac{\kappa_{\mathrm{mag}}(B,T)}{\kappa_{\mathrm{mag}}(0,T)} = \frac{\pi^3\alpha(T) - 2\pi^2 h^2 n_B'(h)}{\pi^3\alpha(T) - (2\pi^2 + 4h^2) h^2 n_B'(h)} \tag{13}$$

with $n_B(h) = 1/(e^h - 1)$. As shown in Fig. 2, the field dependence of the thermal conductivity is predicted to show a pronounced dip at $B \sim T$. For small $\alpha(T) \ll h^2 n_B'(h)$, i.e. weak disorder and not too strong fields,

one finds

$$\frac{\kappa_{\rm mag}(B,T)}{\kappa_{\rm mag}(0,T)} \approx \frac{1}{1+2h^2/\pi^2}, \tag{14}$$

see Fig. 2. This implies a strong reduction of $\kappa_{\rm mag}$ of order 1 for $\mu_B B \sim k_B T$ as long as the renormalized disorder is sufficiently weak, $\alpha(T) \ll 1$. This is the main result of the paper. For large fields or stronger effective disorder one obtains a small suppression of $\kappa_{\rm mag}$,

$$\frac{\kappa_{\rm mag}(B,T)}{\kappa_{\rm mag}(0,T)} \approx 1 - \frac{-4n'_B(h)h^4}{\pi^3\alpha(T)}, \tag{15}$$

giving rise to a minimum at $h \approx 3.8$ of size $0.63/\alpha(T)$.

To allow for a quantitative comparison to the experiment of Ref. [5], one needs to estimate $\alpha(T)$. For this, both the strength of impurity scattering and the strength of the renormalized Umklapp scattering $g(T)$ have to be determined. Fortunately, for simple Heisenberg chains the latter is known analytically from the Bethe Ansatz. Translating our notations to those used in [15], we obtain $g(T) = \tilde{g}(T)\pi^2 Ja/2$ and $\tilde{g}$ is obtained by solving the equation[15]

$$\frac{1}{\tilde{g}} + \frac{\ln(\tilde{g})}{2} = \ln\left(\frac{e^{1/4+\gamma}\sqrt{\pi/2}J}{T}\right) \tag{16}$$

where $\gamma = 0.5772...$ is the Euler constant. Within the precision of our calculation, $\tilde{g} \approx 1/\ln(J/T)$, but we use the more precise formula (16) which includes subleading corrections below. The disorder strength for the sample of CuPzN measured in [5] can in principle be obtained from $\kappa_{\rm mag}(B=0,T)$. Unfortunately, a large phonon background prohibits a direct measurement of this quantity but a crude estimate, $\kappa_{\rm mag} \approx 3.5\,T^2\,{\rm Wm^{-1}K^{-3}}$, can be obtained from the behavior of κ at large fields (see [5] for details). Using this estimate, we find for the heat conductivity per spin chain $\kappa_{\rm mag} \approx 2.1\,10^{-18}\,T^2\,{\rm WmK^{-3}}$. For the four lowest temperatures, $T = 0.37, 0.66, 0.96, 1.48\,{\rm K}$, shown in Fig.1, we obtain from Eq. (12) $\alpha(T) \approx 0.52, 0.12, 0.049, 0.016$, respectively.

These estimates allow a quantitative comparision of theory and experiment. There are two main discrepancies between theory and experiment which can be seen from a direct comparison of Fig. 1 and Fig. 2. First, there is a discrepancy in the position of the minimum (located at $h \approx 3$ in the experiment and at $h \approx 4$ within the theory) and second, the size of the dip of the order of 10% is much smaller than the predicted reduction of more than 50% – or the estimate for α appears to be almost two orders of magnitude too small. What can be the origin of the clear discrepancy? First, one should note that for the temperatures and magnetic fields shown in Fig. 1 both subleading effects of order $\ln(B/T)/\ln(J/T)$ or $\ln(\ln(J/T))/\ln(J/T)$ and band-curvature effects (the overall downturn of $\kappa_{\rm mag}$ in large fields) neglected in our calculation can become important. For example, $\tilde{g}^2$ calculated to leading order is for $J/T = 30$ a factor 2.5 larger than the value obtained from Eq. (16).

More importantly, we believe that our model (2) does not capture all aspects of the physics in the CuPzN samples correctly. Especially, modeling the disorder by Eq. (6) might not be appropriate. This was also the conclusion of Ref. [5] from an analysis of the heat conductivity at large fields $B \sim 15\,{\rm T}$ in the quantum critical regime where the magnetization is close to saturation. Indeed, for other types of disorder the matrix (11) will have a different structure which will affect the quantitative predictions while the qualitative picture will remain unmodified. For example, it might be necessary to take the interplay of forward scattering from impurities and interactions into account. Forward scattering affects transport at $B = 0$ only very weakly but can reduce the Umklapp dip in $\kappa_{\rm mag}$ considerably as the suppression of $\kappa_{\rm mag}$ at larger fields relies on momentum conservation. A more realistic modeling of disorder should also account for the possibility that defects cut the one-dimensional chains in long pieces[16]. In such a situation, one has also to model how phonons (or weak inter-chain interactions) couple heat into and out off such long chain segments[5,16].

Conclusions: Our theoretical calculations show that the heat conductivity of weakly disordered spin chains is very sensitive to moderate magnetic fields $B \sim T$. A pronounced dip in the field dependence of κ arises from the shift of the Fermi surface of the spinons induced by the magnetic field. The effect of Umklapp scattering on the heat conductivity turns out to be strongest when the Fermi surface is shifted from the commensurate position at $B = 0$ by an amount of the order of its thermal broadening.

It is interesting to note that, according to our theory, this effect should be observable for a wide range of parameters including spin-chains which have – in contrast to CuPzN – large exchange couplings of several hundred Kelvin. Due to the strong B dependence, it should be possible to identify dips in $\kappa_{\rm mag}$ even in the presence of a large phonon background. However, for such systems, the effective disorder has to be sufficiently small, $\alpha(T) \lesssim 10$. To obtain an effective disorder strength of the order of 1 at $B \sim T$, typical fluctuations of the exchange coupling have to be of the order of B (for this crude estimate we used $D_{\rm dis} \sim (\delta J)^2 a$ and neglected logarithmic renormalizations) $\delta J/J \lesssim \mu_B B/J$. Furthermore, $\Delta k \sim \mu_B B/(Ja)$ is also small for large J and a necessary condition for the quantitative validity of our calculations is that there is no substantial forward scattering on the associate length scale $1/\Delta k \sim aJ/(\mu_B B)$. In systems with large J and strong phonon scattering, one has also to take into account[10] that the sound velocity c is smaller than the spinon-velocity v. Therefore it may happen that the position of the dip in $\kappa_{\rm mag}$ moves to lower values, $h \sim c/v$, as the relevant energy scale[10] for phonon-assisted Umklapp scattering is $c\Delta k$ rather than $v\Delta k$.

Acknowledgments

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